

**Original citation:**

Bassiri, Riccardo, Hart, Martin, Byer, Robert L, Borisenko, Konstantin B, Evans, Keith, Fejer, Martin M, Lin, Angie C, MacLaren, Ian, Markosyan, Ashot S, Martin, Iain W, Route, Roger K and Rowan, Sheila (2014) Investigating the medium range order in amorphous Ta<sub>2</sub>O<sub>5</sub> coatings. In: Electron Microscopy and Analysis Group Conference 2013 (EMAG2013), University of York, York, UK, 3-6 September 2013. Published in: Journal of Physics: Conference Series, Volume 522

**Permanent WRAP url:**

<http://wrap.warwick.ac.uk/62939>

**Copyright and reuse:**

The Warwick Research Archive Portal (WRAP) makes this work of researchers of the University of Warwick available open access under the following conditions.

This article is made available under the Creative Commons Attribution- 3.0 Unported (CC BY 3.0) license and may be reused according to the conditions of the license. For more details see <http://creativecommons.org/licenses/by/3.0/>

**A note on versions:**

The version presented in WRAP is the published version, or, version of record, and may be cited as it appears here.

For more information, please contact the WRAP Team at: [publications@warwick.ac.uk](mailto:publications@warwick.ac.uk)

warwick**publications**wrap  
  
highlight your research

<http://wrap.warwick.ac.uk/>

## Investigating the medium range order in amorphous Ta<sub>2</sub>O<sub>5</sub> coatings

This content has been downloaded from IOPscience. Please scroll down to see the full text.

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 137.205.202.117

This content was downloaded on 26/08/2014 at 16:03

Please note that [terms and conditions apply](#).

# Investigating the medium range order in amorphous Ta<sub>2</sub>O<sub>5</sub> coatings

**Riccardo Bassiri<sup>1,a</sup>, Martin Hart<sup>2,b</sup>, Robert L. Byer<sup>1</sup>, Konstantin B. Borisenko<sup>3</sup>, Keith Evans<sup>4</sup>, Martin M. Fejer<sup>1</sup>, Angie C. Lin<sup>1</sup>, Ian MacLaren<sup>2</sup>, Ashot S. Markosyan<sup>1</sup>, Iain W. Martin<sup>2</sup>, Roger K. Route<sup>1</sup>, Sheila Rowan<sup>2</sup>**

<sup>1</sup> E.L. Ginzton Laboratory, Stanford University, Stanford, California, USA

<sup>2</sup> SUPA School of Physics and Astronomy, University of Glasgow, Glasgow, UK

<sup>3</sup> Department of Materials, University of Oxford, Oxford, UK

<sup>4</sup> Department of Physics, University of Warwick, Warwick, UK

<sup>a</sup>rbassiri@stanford.edu

<sup>b</sup>m.hart.1@research.gla.ac.uk

**Abstract.** Ion-beam sputtered amorphous heavy metal oxides, such as Ta<sub>2</sub>O<sub>5</sub>, are widely used as the high refractive index layer of highly reflective dielectric coatings. Such coatings are used in the ground based Laser Interferometer Gravitational-wave Observatory (LIGO), in which mechanical loss, directly related to Brownian thermal noise, from the coatings forms an important limit to the sensitivity of the LIGO detector. It has previously been shown that heat-treatment and TiO<sub>2</sub> doping of amorphous Ta<sub>2</sub>O<sub>5</sub> coatings causes significant changes to the levels of mechanical loss measured and is thought to result from changes in the atomic structure. This work aims to find ways to reduce the levels of mechanical loss in the coatings by understanding the atomic structure properties that are responsible for it, and thus helping to increase the LIGO detector sensitivity. Using a combination of Reduced Density Functions (RDFs) from electron diffraction and Fluctuation Electron Microscopy (FEM), we probe the medium range order (in the 2-3 nm range) of these amorphous coatings.

## 1. Introduction

Amorphous heavy metal oxides, such as Ta<sub>2</sub>O<sub>5</sub>, are widely used as the high refractive index layer of highly reflective dielectric coatings. Ion-beam sputtered Ta<sub>2</sub>O<sub>5</sub> is used in the dielectric coatings of high performance applications such as the ground based Laser Interferometer Gravitational-wave Observatory (LIGO). Mechanical loss, which is directly related to Brownian thermal noise from the coatings, forms an important limit to the sensitivity of the LIGO detector [1].

It has previously been shown that heat-treatment and TiO<sub>2</sub> doping of the Ta<sub>2</sub>O<sub>5</sub> coatings can cause noticeable changes to the levels of mechanical loss measured [2, 3]. This is thought to be the result of changes in the atomic structure, and recent observations have shown a direct correlation between the mechanical loss and local atomic structure [4]. Therefore, understanding the atomic structure and properties of these coatings is an area of intensive investigation. We ultimately aim to find ways to reduce the levels of mechanical loss in the coatings, and thus help to increase the LIGO detector sensitivity.

In this work, the atomic structure of heat-treated Ta<sub>2</sub>O<sub>5</sub> coatings is studied and compared to previously measured values of mechanical loss, and the first results of the medium range order from a heat-treated Ta<sub>2</sub>O<sub>5</sub> coating is presented.

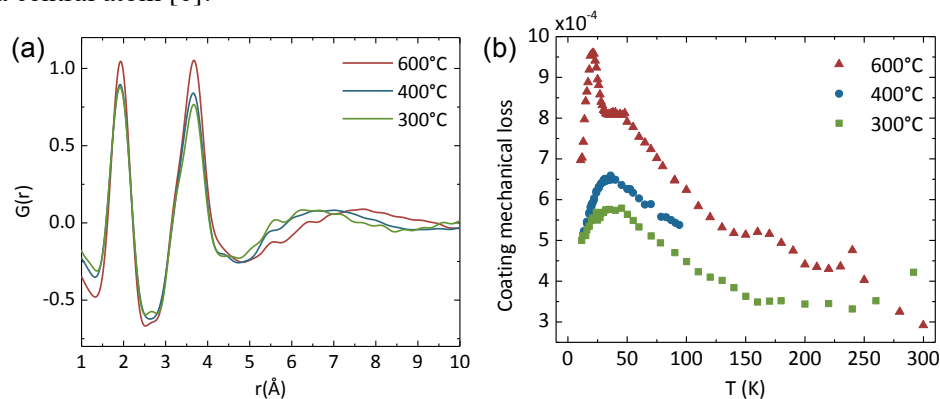


## 2. Experimental details

The coatings studied in this work were 500 nm thick argon ion-beam sputtered Ta<sub>2</sub>O<sub>5</sub> on silica (SiO<sub>2</sub>) glass substrates, manufactured by the Commonwealth Scientific and Industrial Research Organisation (CSIRO, Materials Science and Engineering Division, West Lindfield, NSW, Australia). Heat-treatment of the samples was carried out at 300, 400 and 600°C for a period of 24 hours in air. TEM samples were prepared using a conventional cross section method, finished with argon ion polishing. Full details of the TEM sample preparation technique can be found in a detailed guide prepared by Muller *et al.* [5]. Investigations of the medium range order were carried out on a JEOL ARM 200FCS at 25 cm camera length, using a 2 nm FWHM probe and recording onto an 11 megapixel Gatan Orius SC1000A CCD at 4x binning. The high dynamic range imaging routine used to improve the short range order measurements was provided by Bernhard Schaffer of Gatan and scales and stitches together multiple diffraction patterns taken at a range of exposure times. The FEM data collection entailed formation of the desired probe size, positioning of the probe on a bright field image where a line of measurements could be taken from, thereafter in diffraction mode taking roughly seventy exposures, each two seconds long. The resultant diffraction patterns were then cropped around their common centre and stacked together. Each diffraction pattern was then radially averaged before the variance was computed through the stack.

## 3. Linking atomic structure and mechanical loss

To study the atomic structure of the Ta<sub>2</sub>O<sub>5</sub> coatings, the Short Range Order (SRO) of the material is studied. The SRO is measured by computing the Reduced Density Function (RDF) from radially averaged electron diffraction data. RDFs give a statistical representation of where atoms sit with regards to a central atom [6].



**Figure 1.** (a) Reduced density functions [7], (b) mechanical loss [2] of heat-treated Ta<sub>2</sub>O<sub>5</sub> coatings.

Figure 1(a) contains the RDFs obtained from the diffraction patterns from coatings that have undergone different heat-treatments [7, 8]. The RDFs show subtle changes in SRO, the peaks become taller and narrower, indicating a slight increase in short-range ordering with increasing heat-treatment temperature. Figure 1(b) shows previously measured mechanical loss measurements by Martin *et al.* [2] and highlights the substantially different mechanical losses measured from these coatings, showing low temperature loss peaks which grow with increasing heat-treatment temperature. The substantial changes observed in the temperature dependent mechanical loss, induced by heat-treatment, is interesting in that the atomic structure shows only small and subtle changes in the SRO. It is therefore hypothesised that there may be larger scale changes in the atomic structure responsible for the changes observed in the mechanical loss, and that these are occurring at a longer range than the SRO is sensitive to, i.e., greater than 1 nm.

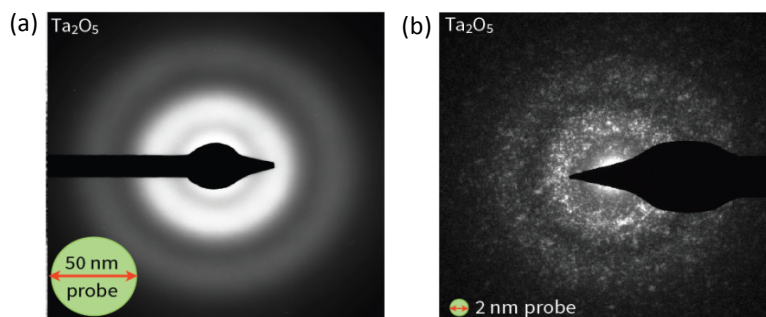
#### 4. Improving the short range order

A recent improvement in RDF measurements has been developed which may help to probe any structural changes in more detail. High Dynamic Range (HDR) diffraction imaging is the process of taking multiple diffraction patterns at different exposure times, and stitching them together to create one image. This increases the SNR at the faintest area of the image (high- $q$ ), whilst maintaining an accurate representation of the centre of the diffraction pattern.

This is an important improvement as it allows the possibility to easily obtain a  $q$ -range of  $25 \text{ \AA}^{-1}$  with  $0.0031 \text{ \AA}^{-1} \text{ pixel}^{-1}$  resolution (highlighted in figure 3(a)), with planned experiments out to a  $50 \text{ \AA}^{-1}$  range. Assuming elastic scattering we define  $q = 4\pi \sin \theta / \lambda$ , with  $2\theta$  being the scattering angle and  $\lambda$  the wavelength of the radiation used. The RDFs in figure 1(a) were computed for data recorded out to  $12 \text{ \AA}^{-1}$ . The new procedure with a doubled  $q$ -range allows much better background fitting which in turn allows better peak representation in the RDF, especially at low  $r$ , and thus improves the reliability of our RDFs. This will be implemented in all future RDF investigations.

#### 5. Probing the medium range order

One way to measure changes in the atomic structure over a longer range is to use Fluctuation Electron Microscopy (FEM). FEM is a diffraction and/or imaging technique that probes the Medium Range Order (MRO), in the roughly 1 to 3 nm range. FEM measures the MRO by measuring intensity fluctuations from nano-volumes in the sample material. These fluctuations are quantified by computing the normalised variance of the diffracted intensity, which is maximally sensitive when the electron probe size is of comparable length scale to the MRO structural ordering being probed. Originally proposed by Gibson and Treacy [9], the technique was initially carried out using dark-field imaging in the TEM. We have used nanodiffraction FEM to collect data, in a similar way to that described by Voyles and Muller's [10].

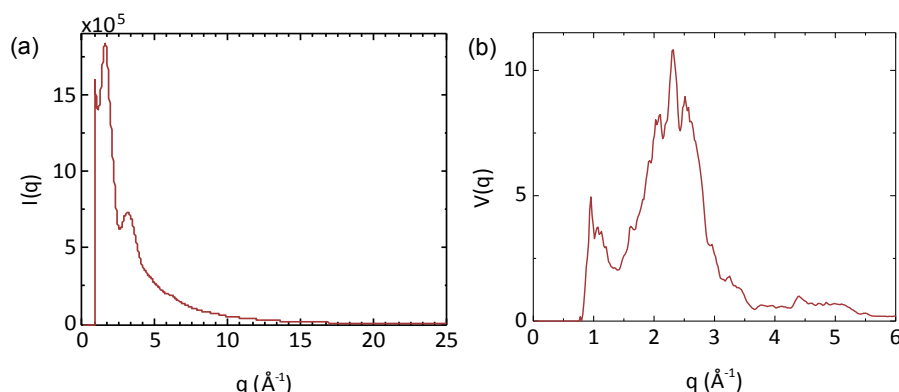


**Figure 2.** (a) Electron diffraction pattern with 50 nm probe, (b) FEM electron diffraction pattern with 2 nm probe from a 600°C heat-treated  $\text{Ta}_2\text{O}_5$  coating.

Figure 2(a) shows the diffuse rings of the electron diffraction pattern obtained from an amorphous  $\text{Ta}_2\text{O}_5$  coating using a 50 nm probe that is used routinely in RDF analysis. Here, any MRO structural detail is washed out by the inherent averaging over the relatively large volume probed. Figure 3(b) shows the FEM diffraction pattern collected from a 600°C heat-treated  $\text{Ta}_2\text{O}_5$  coating, and highlights the presence of spatial fluctuations when using a 2 nm probe. Quantification of these spatial fluctuations is determined by examining the normalized variance of the diffracted intensity,  $V(q)$ .

The 600°C heat-treated  $\text{Ta}_2\text{O}_5$  coating sample was investigated. Figure 3(a) shows the HDR intensity profile from a 50 nm probe (for use in RDF investigations), and figure 3(b) shows the corresponding normalized variance when a 2 nm probe is used, which confirms the presence of MRO.

Planned work in the future will use this method of FEM to study the MRO of  $\text{Ta}_2\text{O}_5$  coatings as a function of heat-treatment temperature. Here it will be expected that heat-treatment causes larger changes in the MRO than are obvious in the SRO, and the aim will be to correlate this to the observed changes in mechanical loss.



**Figure 3.** (a) HDR intensity profile and (b) normalised variance  $V(q)$  plot from a 600°C  $\text{Ta}_2\text{O}_5$  coating.

## 6. Conclusions and future work

Heat-treatment causes subtle changes in the SRO atomic structure of amorphous  $\text{Ta}_2\text{O}_5$  coatings. However, there are substantial changes observed in temperature dependant mechanical loss measurements. Larger changes in the atomic structure, which may more readily explain the observed changes in the mechanical loss, are thought to be occurring in the MRO, and work has begun to probe this using FEM.

Further work will be performed using FEM at varying probe sizes to pin down the length scale of the MRO as described by Voyles *et al.* [10]. We also plan on using FEM, in conjunction with improved high dynamic range RDF data, to create atomic models that include MRO as a key input into the reverse Monte Carlo and molecular dynamics based atomistic simulations.

## Acknowledgements

We would like to thank the support of colleagues in the LIGO Scientific Collaboration, SUPA and the University of Glasgow for the provision of the MagTEM facility used in this work and Bernhard Schaffer of Gatan for the HDR imaging script. We also wish to acknowledge funding from the NSF (Grant No: PHY-1068596) and STFC (Grant No: ST/J000361/1).

## References

- [1] Harry G M, Armandula H, Black E, Crooks DRM, et al. 2010 Appl. Opt. **45** 1569
- [2] Martin I W, Bassiri R, Nawrodt R, Fejer M M, et al. 2010 Class. Quantum Grav. **27** 225020
- [3] Harry G M, Abernathy M R, Becerra-Toledo A E, Armandula H, et al. 2007 Class. Quantum Grav. **24** 405-415
- [4] Bassiri R, Evans K, Borisenko K B, Fejer M M, et al. (2013) Acta Mat. **61** 1070–1077, 2013
- [5] Muller E, Abou-Ras D, 2004 *Preparation of Cross-section Samples for Transmission Electron Microscopy* (Swiss Federal Institute of Technology Zurich)
- [6] Cockayne D J H 2007 Annu. Rev. Mater. Res. **37** 159-87
- [7] Bassiri R 2011 *The Atomic Structure and Properties of Mirror Coatings for use in Gravitational Wave Detectors* (University of Glasgow) Ph.D. Thesis
- [8] Bassiri R, Borisenko K B, Cockayne D J H, Hough J, et al. 2011 App. Phys. Lett. **98** 031904
- [9] Treacy M M J and Gibson J M 1996 Acta Cryst. A **52** 212
- [10] Voyles P M and Muller D A 2002 Ultramicroscopy **93** 147